2版

様　式　C－19、F－19－1、Z－19　（共通）

科学研究費助成事業　　研究成果報告書

令和　６ 年　６ 月 １１ 日現在

機関番号：８２４０１

研究種目：若手研究

研究期間：2022～2023

課題番号：２２Ｋ１４２９３

研究課題名（和文）Development of a Simple and Scalable Method for Organic Semiconductor Single Crystal Growth and Formation of Multi-Single Crystal Thin Films for Applications in Field-Effect Transistor-Based Devices.

研究課題名（英文）Development of a Simple and Scalable Method for Organic Semiconductor Single Crystal Growth and Formation of Multi-Single Crystal Thin Films for Applications in Field-Effect Transistor-Based Devices.

研究代表者

Bulgarevich Kirill（Bulgarevich, Kirill）

国立研究開発法人理化学研究所・創発物性科学研究センター・特別研究員

研究者番号：６０８８０２６８

交付決定額（研究期間全体）：（直接経費）　　3,500,000円

研究成果の概要（和文）：新たな結晶成長手法「間接昇華法」を開発し、真空レス・溶媒レスで有機半導体単結晶を大面積に形成することに成功した。得られた無数のフリースタンディング単結晶を簡易的な押し付け転写とラビングプロセスによりデバイス基板に転写し、小さな結晶が折り重なって大面積をカバーする「多単結晶膜」とした。多単結晶膜を活性層とした有機トランジスタが単結晶デバイスに近い動作を示すことを確認し、これらを組み合わせてCMOS素子のようなインバーター特性を示す擬CMOS素子の作製に成功した。さらに、独自の結晶構造シミュレーションアルゴリズムを使用し、多単結晶膜応用も期待される新たな超高移動度有機半導体材料の開発にも成功した。

研究成果の学術的意義や社会的意義
新たに開発された「間接昇華法」および「多単結晶膜」形成は真空や溶媒を用いずに有機半導体の大面積な結晶性薄膜を形成する手法である。特に我々が最近報告した有機半導体MT-ピレンは多単結晶膜トランジスタで(>15 cm2 V-1 s-1)の非常に高い移動度を示し、この技術により環境への影響を抑えた高性能な有機デバイスの実現が期待される。さらに、我々が独自で開発した結晶構造シミュレーションアルゴリズムを使用することでMT-ピレンと同等な移動度を示す新規材料が開発された。この手法は多単結晶膜応用を念頭に置いた溶解性にとらわれない材料設計と構造シミュレーションによる効率的な高移動度新規材料開発につながる。

研究成果の概要（英文）：We developed a novel crystal growth technique named "indirect sublimation", and successfully formed organic semiconductor single-crystals on large-area without the use of vacuum or solvents. These numerous freestanding single-crystals were transferred onto device substrates through a simple press-transfer and rubbing process, creating a so-called "multi-single-crystal (MSC)" film, where small crystals overlap to cover large areas. We confirmed that organic field-effect transistors (OFETs) using this MSC film as the active layer perform similarly to single-crystal devices. By combining such OFETs, we successfully fabricated pseudo-CMOS devices that exhibit inverter characteristics similar to CMOS devices. Furthermore, using our unique crystal structure simulation algorithm, we successfully developed a novel ultra-high mobility organic semiconductor material, which also has potential for MSC film applications.

研究分野：有機半導体

キーワード：有機半導体　有機トランジスタ　単結晶　薄膜　多単結晶膜　大面積　間接昇華法

１．研究開始当初の背景

Organic field-effect transistors (OFETs) are garnering considerable interest for their potential electronic device applications. Single crystals (SC) of organic semiconductors are particularly significant, not only for providing insights into the fundamental properties of these materials but also for enhancing device performance due to their highly ordered crystalline structures. However, SCs require tedious handcrafting for each OFET, involving the precise placement of a separately grown organic semiconductor SC onto a device's electrodes, or the reverse. Moreover, the production of SCs generally involves complex processes, necessitating expensive equipment for vacuum conditions and extended processing times. Therefore, for many practical applications, flexible thin films of organic semiconductors, capable of being deployed over large areas, are favored over SCs. Such printable electronics consume large amounts of solvent (1000 to 10000 molar ratio to organic semiconductor) which often contain halogens. These solvents pose significant environmental concerns due to their deleterious effects.

We have recently developed a novel ultra-high mobility (> 30 cm^2 V^{-1} s^{-1}) material 1,3,6,8-Tetrakis (methylthio)pyrene (MT-pyrene). Unfortunately, solubility of this material in common organic solvents was too low to attempt thin-film formation by solution methods. During fabrication of SC-OFETs by lamination of physical vapor transport (PVT) grown thin-plate SCs of MT-pyrene onto a device substrate and painting source / drain electrodes with colloidal graphite, we have noticed an interesting phenomenon. Placing such thin SCs on top of each other and extending SCs in a chain-manner did not result in deterioration of device properties, and the resulting "chained-SC" OFET operated similarly to an SC-OFET (Fig. 1). Moreover, covering an area of the substrate with SCs entirely also operated as a single OFET. This led to the idea that thin-film-like applications for SCs are possible without use of solvents by developing a method to produce so called "multi-single-crystal (MSC)" films of organic semiconductors.

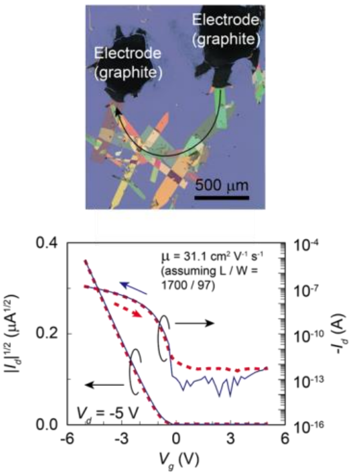


Figure 1. "Chained-single-crystals" OFET of MT-pyrene.

２．研究の目的

The purpose of the proposed research was to develop a practical (simple, scalable, solvent-free, and vacuum-free) method to fabricate SC OFET based devices such as pseudo-CMOS inverters. For this purpose, it was proposed to develop a scalable method for crystal growth from vapor phase, and their easy transfer on the device substrate for MSC film formation. Another important purpose of this research was to develop novel ultra-high mobility materials potentially suitable MSC film applications.

３．研究の方法

The research methods could be divided into three parts: development of simple and scalable method for crystal growth and MSC film formation; fabrication of MSC-OFETs and MSC-OFET-based devices; development of novel ultra-high mobility materials.

(1) Crystal growth and formation of MSC films.

The problem of poor scalability of single-crystal growth was solved by developing a novel crystal growth method named "indirect sublimation" (Fig 2a). In this method, the source material is placed on a heater covered by a glass plate (blocking substrate) and also covered by a Petri dish. The target substrate is placed on the top inside of the Petri dish. The sublimated source material escapes from the gap under the blocking substrate and forms crystals on the above target substrate. The process is performed under atmospheric pressure (in air for some materials). This method is based on a recently reported microspacing in-air sublimation (MSS) method [*Chem. Mater.* **30**, 412 (2018); *Nature Commun.* **6**, 5954 (2014); *Chem. Mater.* **31**, 6696 (2019)], but introduces a blocking substrate between the source and the target substrates and places the whole setup in a semi-confined space. The blocking substrate is important for preventing the source powder particles from directly reaching the target substrate (hence the naming of "indirect sublimation"). The semi-hermetical nature

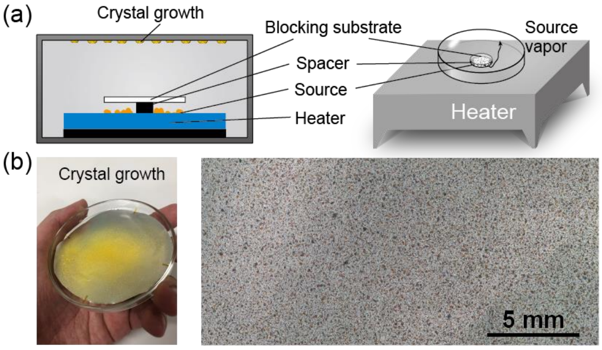


Figure 2. (a) Indirect sublimation setup under atmospheric pressure. (b) Crystals grown by indirect sublimation.

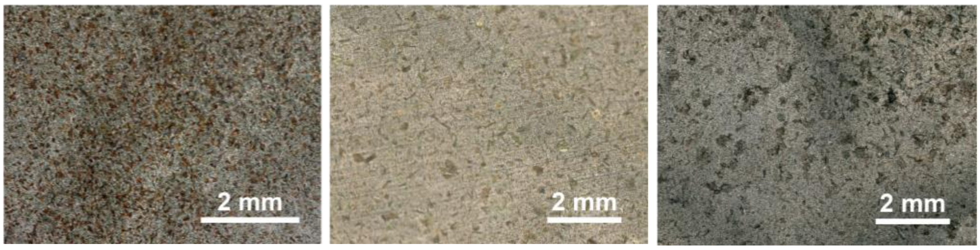


Figure 3. Results for indirect sublimation of rubrene (left), (b) DNTT (middle), and CYHEX-NDI (right).

of the space below the Petri dish ensures that almost no material escapes outside, and all are used to grow crystals on the surface of the lowest temperature. By using this method, remarkably uniform and dense growth of free-standing crystals can be achieved on a large (scalable) area almost independent of initial material distribution (Fig. 2b). It was confirmed that the crystal growth by indirect sublimation is possible for MT-pyrene, rubrene, DNTT, N,N′-bis(cyclohexyl)naphthalene diimide (CYHEX-NDI) (Fig.3), 1,3,6,8-Tetrakis (methylseleno)pyrene (MS-pyrene), pentacene, and others.

The crystals grown by indirect sublimation are free-standing and still it was not practical to use them for OFET applications. The conventional method for fabricating single-crystal OFETs is to take one crystal manually using a tungsten needle and laminate the crystal on the device substrate (usually Si substrate with modified SiO_2 layer). Then, the source and gate electrodes are formed on each end of the laminated crystal by using physical vapor deposition and a shadow mask or by painting the colloidal graphite or gold paste. Since the target substrate in indirect sublimation is a large flat surface, it was found that the lamination process can be largely simplified using press-transfer onto CYTOP-coated SiO_2/Si substrate (Fig4 a). The simple press-transfer of the resulting crystals onto a target substrate resulted in a uniform coverage by an MSC film, but the resulting devices showed poor on/off ratio. This was probably because some of the crystals were not completely attached to the substrate or to crystals below resulting in gate voltage not being applied. This problem was solved by introducing a rubbing machine to push down / remove the excess crystals to / from the substrate (Fig. 4b). Such process resulted in a film of interconnecting and overlapping SC thin plates (< 0.05 mm^2 each) covering a relatively large substrate surface (~ 1 cm^2).

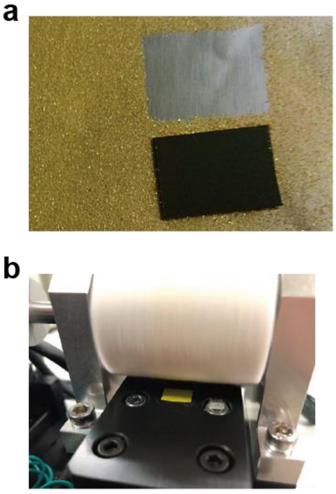


Figure 4. Formation of multi-single-crystal film. (a) The device substrate is pressed against crystals grown on Al substrate (top) and then moved onto different section of crystals (bottom). Note almost all crystals are removed from Al by pressing. (b) Rubbing process.

(2) MSC-OFETs and MSC-OFET-based devices.

OFETs with MSC film of MT-pyrene as an active layer were completed by painting source / drain electrodes with colloidal graphite. The OFETs showed almost ideal device characteristics with on/off ratio of > 10^6, and high mobility (> 15 cm^2 V^{-1} s^{-1}) at < 10 V operation, comparable to single-crystal devices (Fig 5a). CYHEX-NDI was tested to produce an n-type MSC film for complementary circuit application with MT-pyrene. Unfortunately, the carrier mobility of CYHEX-NDI was largely reduced in the MSC form from ~0.5 cm^2 V^{-1} s^{-1} down to ~0.03 cm^2 V^{-1} s^{-1}. Although the exact mechanism of carrier transport in MSC films is currently unknown and has to be investigated in more detail, this is probably related to lack of out-of-plane carrier transport in CYHEX-NDI. Both MT-pyrene and CYHEX-NDI crystallize into brickwork structures in which the carrier transport is dominated by the two π-stacking directions which both lie in the substrate surface plane. However, for carrier transport between the crystals in the MSC film which lie on top of

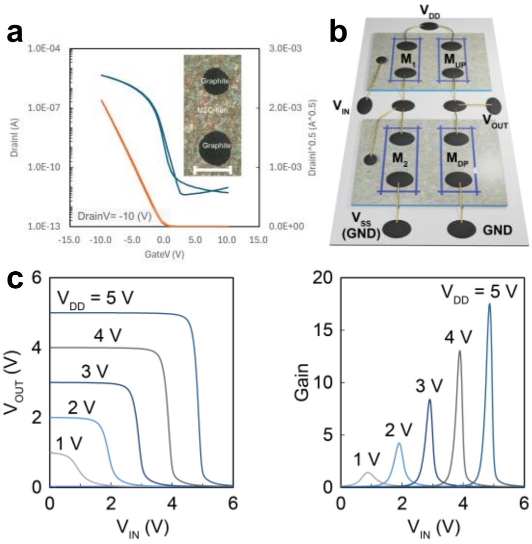


Figure 5. (a) MT-pyrene MSC OFET showing mobility of 16.0 cm^2 V^{-1} s^{-1}. The inset is an optical microscope image of the OFET (scale bar is 0.5 mm). (b) Schematic of the wiring of OFETs to produce a pseudo-CMOS inverter. (c) Operation of MT-pyrene pseudo-CMOS inverter.

each other, probably charge transport in the thickness direction of the crystals must occur. Such charge transport is possible to some extent in MT-pyrene due to molecule side-to-side interactions but is blocked in CYHEX-NDI by the bulky cyclohexyl groups. A suitable n-type material for MSC film applications was not found yet.

It is possible to fabricate a pseudo-CMOS inverter using four p-type OFETs (Fig. 5b). MT-pyrene pseudo-CMOS inverter fabricated using graphite paste and gold wiring showed good inverter characteristics and relatively good gain of ~20 at 5V (Fig. 5c). Similar characteristics were obtained for MT-pyrene pseudo-CMOS inverter made using OFETs with MoO_x/Au electrodes (Fig. 6).

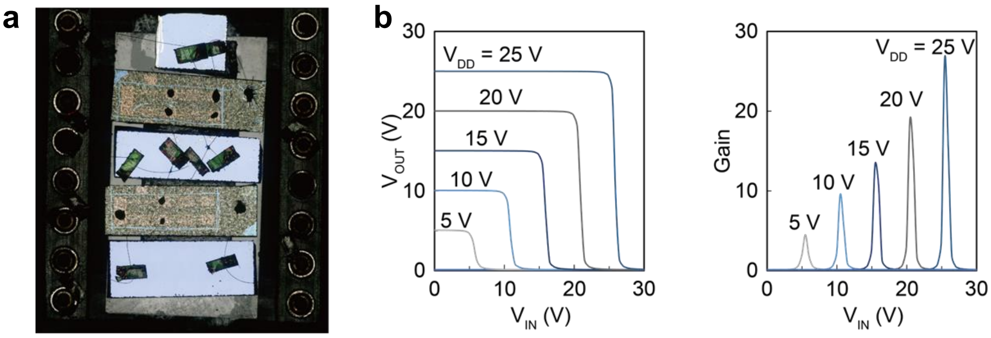


Figure 6. (a) Optical microscope image and (b) characteristics of a pseudo-CMOS inverter composed of four MT-pyrene MSC OFETs with MoOx/Au source/drain electrodes.

(3) Development of novel ultra-high mobility materials.

The main material used throughout this research, MT-pyrene, shows ultra-high mobility (> 30 cm^2 V^{-1} s^{-1}) in SC-OFETs and very high mobility of > 10 cm^2 V^{-1} s^{-1} even in MSC OFETs. On the other hand, MS-pyrene which is selenium substitution of MT-pyrene, despite having almost identical brickwork crystal structure (Fig. 6), shows only moderately high mobility of ~ 7 cm^2 V^{-1} s^{-1} in SC-OFETs. This is because the small differences between the crystal structures of MT- and MS-pyrene result in large difference in intermolecular orbital overlaps. This means that choosing the molecules that crystallize into promising crystal structures is not enough to reliably realize high carrier mobility.

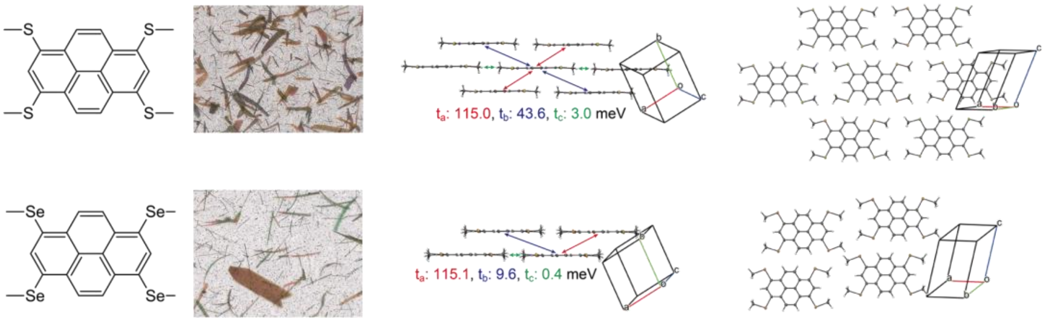


Figure 6. Molecular structures, optical microscope images, and crystal structures of MT-pyrene (top) and MS-pyrene (bottom). The transfer integrals in the brickwork layers are indicated as arrows.

To tackle this issue, the applicant has developed a novel algorithm for brickwork crystal structure simulation named "in silico crystallization (ISC)". This algorithm is drastically different from the conventional crystal structure prediction relying on random search in vast space. Rather, ISC simulates the brickwork crystal structure (one target crystal structure type) using step-by-step optimization of bimolecular interactions. By focusing on face-to-face (F), side-to-side (S), and end-to-end (E) directions of molecules, three vectors that define the crystal structure are found by minimizing the sum of interaction energies of certain molecular pairs (Fig. 7). This is possible because the intermolecular interactions in the brickwork structure are dominated by π-π (F) interactions followed by S and E, which enables optimization of π-stacking position without considering S and E interactions. Using this approach, the subtle differences between the crystal structures of MT- and MS-pyrene were successfully simulated explaining the differences between their carrier mobility.

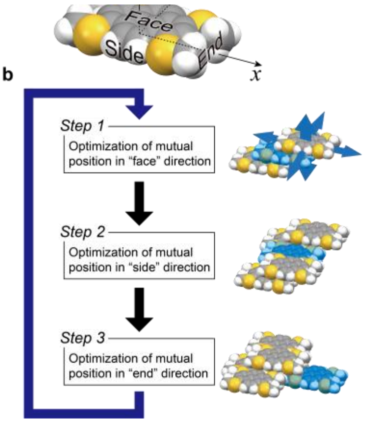
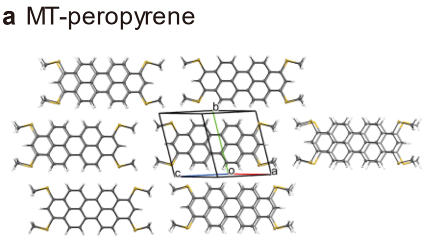


Figure 7. (a) Definition of face, side, and end directions of a molecule. (b) Flow of crystal structure simulation of "in silico" crystallization.

ISC was applied to unknown molecules with similar methylthiolation, which were expected to crystallize into brickwork structures: 2,5,8,11-tetrakis(methylthio)perylene (MT-perylene), 1,3,8,10-tetrakis(methylthio)peropyrene (MT-peropyrene), and 2,5,10,13-tetrakis(methythio)terrylene (MT-terrylene). It was found that MT-perylene and MT-terrylene were not promising as organic semiconductors despite having a larger π-cores than MT-pyrene. On the other hand, MT-peropyrene was expected to show high carrier mobility due to large transfer integrals in two π-stacking directions of the simulated crystal structure. These simulation results were then confirmed experimentally by performing the syntheses of MT-perylene and MT-peropyrene. The experimental crystal structures of both materials were basically identical to the results of the simulation, and experimentally evaluated carrier mobilities were 0.2 and 30 $cm^2 V^{-1} s^{-1}$ for MT-perylene and MT-peropyrene, respectively. Thus, another example of ultrahigh mobility material following MT-pyrene, MT-peropyrene, was successfully found using a combination of crystal structure “manipulation” and crystal structure simulation (Fig. 8).



b

	Simulated	Experimental[a]
Crystal System	*triclinic*	*triclinic*
Space Group	(P1)[b]	P–1
a /Å	6.67	6.6732(2)
b /Å	8.85	8.9016(4)
c /Å	10.23	10.0433(3)
α /°	89.5	90.525(3)
β /°	104.5	103.787(3)
γ /°	108.9	108.291()
V /Å^3	551.7	547.85(4)
E_{int}(15) /kcal mol	−131.2	−138.8
RMSD15 /Å	0.34	–
Comment	–	This work

[a] Data obtained at 100 K.

[b] Space group was not explicitly determined.

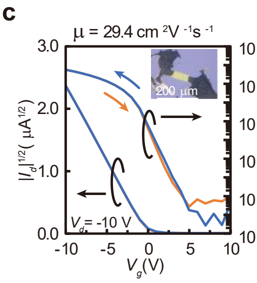


Figure 8. MT-peropyrene (a) experimental structure shown as an overlay on top of the structure simulated by ISC. (b) Comparison of cell parameters of crystal structure simulation and real structure. (c) OFET characteristics of MT-peropyrene

Unfortunately, the MSC film formation of MT-peropyrene could not be achieved during this research period. This is because of difficulties of synthesis and handling of the material. Sublimation temperature of MT-peropyrene is higher than its decomposition temperature under atmospheric pressure because of large molecule size. On the other hand, indirect sublimation method requires some degree of convection flow for plate-like crystal growth. Thus, preside condition optimization of temperature and pressure are required for which we would need to synthesize larger amount of MT-peropyrene. I will continue evaluation of this promising material in the future. In addition, ISC yielded many more promising novel material candidates to synthesize for achieving high mobility in SC and MSC applications.

４．研究成果

A novel method for simple, scalable, solution-free, and vacuum-free crystal growth named “indirect sublimation” was developed. The crystal growth achieved by indirect sublimation was almost completely independent of initial material distribution and achievable on large flat surfaces. Such crystal growth nature enabled development of a simple press transfer method to produce a so-called “multi-single-crystal (MSC)” film of interconnecting and overlapping crystals. MSC OFETs of MT-pyrene showed almost ideal device characteristics and high carrier mobility (> 15 $cm^2 V^{-1} s^{-1}$)comparable to single-crystal devices (30 $cm^2 V^{-1} s^{-1}$). Pseudo CMOS inverters fabricated using four MT-pyrene MSC OFETs showed good inverter characteristics demonstrating that MSC film-based devices can have similar applications to thin-film-based ones. A novel crystal structure simulation algorithm, “in silico crystallization (ISC)” was developed for aiding the search for high mobility organic semiconductor materials. A novel ultrahigh mobility (>30 $cm^2 V^{-1} s^{-1}$) material, MT-peropyrene which is also promising for MSC film applications, was found with the help of crystal structure simulation.

５．主な発表論文等

〔雑誌論文〕　計8件（うち査読付論文　8件／うち国際共著　0件／うちオープンアクセス　4件）

１．著者名 Takimiya Kazuo、Bulgarevich Kirill、Horiuchi Shingo、Sato Aoi、Kawabata Kohsuke	４．巻 4
２．論文標題 Bandlike versus Temperature-Independent Carrier Transport in Isomeric Diphenyldinaphtho[2,3-<i>b</i>:2′,3′-<i>f</i>]thieno[3,2-<i>b</i>]thiophenes	５．発行年 2022年
３．雑誌名 ACS Materials Letters	６．最初と最後の頁 675～681
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acsmaterialslett.2c00084	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

１．著者名 Bulgarevich Kirill、Horiuchi Shingo、Ogaki Takuya、Takimiya Kazuo	４．巻 34
２．論文標題 1,3,6,8-Tetrakis(methylchalcogeno)pyrenes: Effects of Chalcogen Atoms on the Crystal Structure and Transport Properties	５．発行年 2022年
３．雑誌名 Chemistry of Materials	６．最初と最後の頁 6606～6616
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.chemmater.2c01544	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

１．著者名 Takimiya Kazuo、Bulgarevich Kirill、Sahara Kamon、Kanazawa Kiseki、Takenaka Hiroyuki、Kawabata Kohsuke	４．巻 40
２．論文標題 What Defines a Crystal Structure? Effects of Chalcogen Atoms in 3,<scp>7‐Bis</scp>(methylchalcogeno)benzo[1,2‐<i>b</i>:4,5‐<i>b</i>′]<scp>dichalcogenophene‐Based</scp> Organic Semiconductors[†]	５．発行年 2022年
３．雑誌名 Chinese Journal of Chemistry	６．最初と最後の頁 2546～2558
掲載論文のDOI（デジタルオブジェクト識別子） 10.1002/cjoc.202200302	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

１．著者名 Kanazawa Kiseki、Bulgarevich Kirill、Kawabata Kohsuke、Takimiya Kazuo	４．巻 35
２．論文標題 Uncovered Effects of thieno[2,3-<i>b</i>]thiophene Substructure in a Tetrathienoacene Backbone: Reorganization Energy and Intermolecular Interaction	５．発行年 2022年
３．雑誌名 Chemistry of Materials	６．最初と最後の頁 280～288
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.chemmater.2c03160	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

１．著者名 Bulgarevich Kirill、Horiuchi Shingo、Takimiya Kazuo	４．巻 35
２．論文標題 Crystal‐Structure Simulation of Methylthiolated <i>Peri</i>‐Condensed Polycyclic Aromatic Hydrocarbons for Identifying Promising Molecular Semiconductors: Discovery of 1,3,8,10‐tetrakis(methylthio)peropyrene Showing Ultrahigh Mobility	５．発行年 2023年
３．雑誌名 Advanced Materials	６．最初と最後の頁 2305548 (1-7)
掲載論文のDOI（デジタルオブジェクト識別子） 10.1002/adma.202305548	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

１．著者名 Takimiya Kazuo、Bulgarevich Kirill、Horiuchi Singo	４．巻 11
２．論文標題 Contrasted behaviours of methylthiolated perylene and pyrene as organic semiconductors: implications of molecular electronic structure and crystal structure	５．発行年 2023年
３．雑誌名 Journal of Materials Chemistry C	６．最初と最後の頁 10809～10815
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/d3tc02063k	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

１．著者名 Bulgarevich Kirill、Takimiya Kazuo	４．巻 10
２．論文標題 Crystal-structure simulation of molecular semiconductors: brickwork-related crystal structures of methylthiolated <i>peri</i>-condensed polycyclic aromatic hydrocarbons	５．発行年 2023年
３．雑誌名 Materials Horizons	６．最初と最後の頁 5492～5499
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/d3mh01055d	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

１．著者名 Takimiya Kazuo、Bulgarevich Kirill、Kawabata Kohsuke	４．巻 57
２．論文標題 Crystal-Structure Control of Molecular Semiconductors by Methylthiolation: Toward Ultrahigh Mobility	５．発行年 2024年
３．雑誌名 Accounts of Chemical Research	６．最初と最後の頁 884～894
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.accounts.3c00756	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

〔学会発表〕　計8件（うち招待講演　0件／うち国際学会　2件）

１．発表者名
Bulgarevich Dmitrievich Kiril

２．発表標題
In-silico crystallization: メチルカルコゲノピレンのbrickwork型構造の計算による再現

３．学会等名
第83回応用物理学会秋学術講演会

４．発表年
2022年

１．発表者名
Kirill Bulgarevich

２．発表標題
高移動度有機半導体材料探索：構造-物性相関と結晶構造予測

３．学会等名
第16回物性科学領域横断研究会

４．発表年
2022年

１．発表者名
Bulgarevich Dmitrievich Kirill

２．発表標題
In-silico crystallization (2): brickwork型構造シミュレー ションの実験的確認と適用可能結晶系範囲の拡張

３．学会等名
第70回応用物理学会春学術講演会

４．発表年
2023年

１．発表者名
Kirill Bulgarevich and Kazuo Takimiya

２．発表標題
In silico crystallization: simulation of brickwork-related crystal structures using step-by-step optimization of intermolecular interactions

３．学会等名
CEMS Topical Meeting on Chemistry of π-Conjugated Materials

４．発表年
2023年

１．発表者名
Kirill Bulgarevich and Kazuo Takimiya

２．発表標題
In-silico crystallization (3): pitched pi-積層とinclined brickwork 構造の類似性とシミュレーション

３．学会等名
第84回応用物理学会秋季学術講演会

４．発表年
2023年

１．発表者名
Kirill Bulgarevich and Kazuo Takimiya

２．発表標題
Crystal Growth by “Indirect Sublimation” and Formation of Multi-Single-Crystal Films for Practical Applications of Single-Crystal Organic Semiconductors

３．学会等名
The 7th Symposium for the Core Research Clusters for Materials Science and Spintronics and the 6th Symposium on International Joint Graduate Program in Materials Science and Spintronics（国際学会）

４．発表年
2023年

１．発表者名
Kirill Bulgarevich and Kazuo Takimiya

２．発表標題
Multi-Single-Crystal Films: Towards Practical Applications of Single-Crystal Organic Semiconductors

３．学会等名
CEMS International Symposium on Supramolecular Chemistry and Functional Materials 2024（国際学会）

４．発表年
2024年

１．発表者名
ブルガレビッチ キリル，瀧宮和男

２．発表標題
有機半導体「多単結晶膜」：単結晶デバイスの大面積化

３．学会等名
第71回応用物理学会春季学術講演会

４．発表年
2024年

〔図書〕　計1件

1．著者名 Kirill Bulgarevich, 瀧宮 和男	4．発行年 2024年
2．出版社 シーエムシー出版	5．総ページ数 11
3．書名 有機半導体の開発と最新動向 ～ 第８章　Brickwork型結晶構造のシミュレーションに基づく高移動度有機半導体探索	

〔産業財産権〕

〔その他〕

－

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関